



Adsorption and precipitation of cadmium affected by chemical form and addition rate of phosphate in soils having different levels of cadmium

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HIGHLIGHTS

- We tried to determine the exact mechanism of Cd immobilization in various conditions.
- Immobilization of Cd with P might be attributed to Cd adsorption in low Cd levels.
- Precipitation of Cd might be a dominant mechanism to immobilize Cd in high Cd levels.

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ABSTRACT

Although a number of studies have examined cadmium (Cd) immobilization by phosphate (P) in soils, determining the exact mechanism of Cd immobilization in various conditions, including P chemical form, P rate, and inherent Cd concentrations in soil has not been examined. The objective of this study was to determine changes in Cd immobilization through adsorption and precipitation in different conditions. Arable soils were spiked with inorganic Cd to give a total Cd concentration of 10, 100, and 1000 mg Cd kg⁻¹. K₂HPO₄ (DPP) and KH₂PO₄ (MPP) were selected and mixed with the pretreated arable soil at the rates of 0, 800, 1600 and 3200 mg P kg⁻¹. In soils with 10 and 100 mg Cd kg⁻¹, both P materials similarly increased negative charge of soil and decreased extractable Cd concentration. Using X-ray diffraction (XRD), a diffraction peak revealing the presence of Cd(H₂PO₄)₂ was observed in soil with 1000 mg Cd kg⁻¹ and 3200 mg P kg⁻¹ soil. In addition, X-ray photoelectron spectroscopy (XPS) analysis and modeling for saturation index for Cd minerals proved that formation of CdCO₃ and Cd₃(PO₄)₂ occurred in soil having 1000 mg Cd kg⁻¹ and addition of both DPP and MPP. Immobilization of Cd might be attributed to Cd adsorption in soil with relatively low Cd levels (<100 mg kg⁻¹). Precipitation of Cd(H₂PO₄)₂, CdCO₃, and Cd-phosphate might be a dominant mechanism to immobilize Cd, besides Cd adsorption, in soil with relatively high Cd levels (1000 mg kg⁻¹).

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1. Introduction

Cadmium (Cd) may adversely affect soil biological activity, plant metabolism, and the health of humans and the animal kingdom (IPCS, 1992; Patrick, 2003; Lane et al., 2015). It can be particularly toxic to humans. Itai-itai disease ("it hurts-it hurts disease") is

caused by Cd poisoning (Ikeda et al., 2000; Watanabe et al., 2000), and chronic exposure to Cd can cause nephrotoxicity in humans, mainly due to abnormalities in tubular re-absorption (Kabata-Pendias, 1993; EFSA, 2009; ATSDR, 2012; Ikeda et al., 2015). Plants may contain Cd when it presents in the soil, because it is somewhat mobile in soil (McLaughlin and Singh, 1999; Park et al., 2012; North et al., 2017). Humans can be readily exposed to Cd through food chain by consuming agricultural products and drinking water containing this metal.

The mobilization of metals in soils for plant uptake and leaching

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to groundwater can be minimized by reducing their bioavailability through chemical and biological immobilization (Adriano, 2001). Application of phosphate (P) to Cd contaminated soils is a widely used method to reduce Cd bioavailability. A number of studies have been conducted on Cd immobilization with P materials in soil (Table 1). These studies have reported that P-induced immobilization of Cd in soils could be attributed to two primary factors: (1) precipitation of a variety of Cd crystals, such as $\text{Cd}_3(\text{PO}_4)_2$ and CdCO_3 (Boisson et al., 1999; McGowen et al., 2001; Hong et al., 2014; Kim et al., 2015); and (2) Cd^{2+} adsorption to soil particles induced by an increase in pH and negative charge of soil (Bolan et al., 2003; Wang et al., 2008; Hong et al., 2010). However, several studies simply evaluated the effect of P on Cd immobilization, but did not clearly address the mechanism P promotes Cd immobilization (Chen et al., 2000, 2007; Basta et al., 2001; Seaman et al., 2001; Dheri et al., 2007; Lee and Hong, 2016).

From the review to previous works (Table 1), we assumed that three factors including chemical form of P, addition rate of P, and inherent Cd level in soil could govern adsorption and precipitation of Cd in soil.

Phosphate materials shown in Table 1 are inorganic P compounds formed by ionic bonds between a cation and phosphoric acid (H_2PO_4^- or HPO_4^{2-}). Forms of inorganic P, such as H_2PO_4^- and HPO_4^{2-} affect changes in system pH differently (Lee and Doolittle, 2002). Soil pH is one important factor controlling the mobility of Cd (Bolan et al., 2003; Hong et al., 2010, 2014). Cadmium adsorption increases in elevated pH and negative charge due to the ligand-exchange (i.e., OH^- ions) by P adsorption reaction in P amended soil (Hong et al., 2008). Hong et al. (2014) reported that P induced Cd immobilization could be primarily attributed to Cd adsorption in soil pH below 8.0, while Cd precipitation as CdCO_3 occurred frequently when soil pH was above 8.0.

Some studies have reported that Cd^{2+} adsorption had a greater effect than Cd precipitation in soils with inherently low concentrations of Cd ($<10 \text{ mg Cd kg}^{-1}$) (Bolan et al., 2003; Wang et al., 2008; Hong et al., 2010). However, McGowen et al. (2001) reported that Cd solubility was limited by precipitation of $\text{Cd}_3(\text{PO}_4)_2$

and CdCO_3 mineral with addition of $(\text{NH}_4)_2\text{HPO}_4$ in a smelter-contaminated soil containing a high concentration Cd (1090 mg kg^{-1}).

Some studies have reported that Cd can be precipitated as $\text{Cd}_3(\text{PO}_4)_2$ or CdCO_3 by addition of relatively high quantities of P ($>1600 \text{ mg P kg}^{-1}$) in soils with inherently low concentration of Cd ($<10 \text{ mg Cd kg}^{-1}$) (Boisson et al., 1999; Kim et al., 2015). However, other studies have reported that primary mechanism for Cd immobilization with addition of relatively low quantities of P ($<1600 \text{ mg P kg}^{-1}$) was adsorption of Cd by increase in pH and negative charge in soils with inherently low concentration of Cd (Bolan et al., 2003; Hong et al., 2010).

Although a number of studies have examined Cd immobilization by P in soils, few studies have been conducted on organizing change of mechanism of Cd immobilization in various conditions in detail. Therefore, the objective of this study was to determine changes of Cd immobilization such as adsorption and precipitation in differently given conditions (form of inorganic P, addition rate of P, and Cd concentration in soil).

2. Materials and methods

2.1. Soil characteristics and soil spiking

An upland soil was selected for this study from an experimental field at Pusan National University ($35^\circ 30' 07'' \text{ N}$ and $128^\circ 43' 16'' \text{ E}$) Miryang, South Korea. This soil belongs to the Yongji series (fine loamy, mesic family of Fluvaquentic Eutrudepts). A typical sandy loam soil from the 0–15 cm layer was collected, air-dried, and ground through a 2 mm sieve for analyzing chemical properties (Table 2). The soil was artificially contaminated with inorganic Cd (as Cadmium Chloride, 2.5-Hydrate) at the rates of 10, 100, and 1000 mg kg^{-1} on a dry weight basis. Ten l of distilled water containing 0.4063, 4.063, and 40.63 g of $\text{CdCl}_2 \cdot 2.5\text{H}_2\text{O}$ was mixed with 20 kg arable soil to make concentrations corresponding to 10, 100, and $1000 \text{ mg Cd kg}^{-1}$. It was required to give aging periods for Cd to distribute as various fractions to soil, because Cd in solution of

Table 1
Selected references regarding phosphate-affected mechanisms of cadmium immobilization.

Primary mechanism for Cd immobilization	Phosphate material	Phosphate addition rate	Cadmium concentration in soil	Reference
Adsorption of Cd by increase in pH and negative charge	KH_2PO_4	1000 mg kg^{-1}	10 mg Cd kg^{-1}	Bolan et al. (2003)
Adsorption	Phosphate rock, calcium magnesium phosphate, and single superphosphate	500 g/m^{-2}	5.81 mg kg^{-1}	Wang et al. (2008)
Adsorption of Cd by increase in pH and negative charge	Fused phosphate, fused and superphosphate, rock phosphate, $\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot \text{H}_2\text{O}$, $(\text{NH}_4)_2\text{HPO}_4$, KH_2PO_4 , and K_2HPO_4	1600 mg kg^{-1}	5.57 mg kg^{-1}	Hong et al. (2010)
Precipitation (substitution of Ca in hydroxyapatite by Cd during recrystallization)	Hydroxyapatite	$15,000 \text{ mg kg}^{-1}$	6.0 mg kg^{-1}	Boisson et al. (1999)
Precipitation of $\text{Cd}_3(\text{PO}_4)_2$ and CdCO_3	$(\text{NH}_4)_2\text{HPO}_4$	2300 mg kg^{-1}	1090 mg kg^{-1}	McGowen et al. (2001)
Precipitation of Cd as CdCO_3 in elevated soil pH condition	K_2HPO_4	1600 mg kg^{-1}	5.57 mg kg^{-1}	Hong et al. (2014)
Precipitation of $\text{Cd}_3(\text{PO}_4)_2$ and CdCO_3	K_2HPO_4	$16,000 \text{ mg kg}^{-1}$	5.57 mg kg^{-1}	Kim et al. (2015)
Not clear	Calcium phosphate	20 mg kg^{-1}	18.6 mg kg^{-1}	Chen et al. (2000)
Not clear (probably co-precipitation and surface complexation)	Rock phosphate	$100,000 \text{ mg kg}^{-1}$	295 mg kg^{-1}	Basta et al. (2001)
Not clear (probably precipitation and adsorption by change of soil pH)	Hydroxyapatite	$50,000 \text{ mg kg}^{-1}$	1.1 mg kg^{-1}	Seaman et al. (2001)
Not clear (probably adsorption, precipitation, formation of Ca-Cd-phosphate, and ion exchange)	KH_2PO_4	24 mg kg^{-1}	60 mg kg^{-1}	Dheri et al. (2007)
Not clear (probably co-precipitation of insoluble Cd phosphate mineral)	Hydroxyapatite, phosphate rock, triple-superphosphate, and $(\text{NH}_4)_2\text{HPO}_4$	$1,091 \text{ mg kg}^{-1}$	1.5 mg kg^{-1}	Chen et al. (2007)

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